

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF

JUNJI OTANI ET AL.

APPLICATION NO: 09/657,788

FILED: SEPTEMBER 8, 2000

FOR: ELECTROLUMINESCENT DEVICES COMPRISING  
DIKETOPYRROLOPYRROLES



GROUP ART UNIT: 1614

EXAMINER: M. YAMNITZKY

Commissioner of Patents and Trademarks  
Washington, D.C. 20231

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Sir:

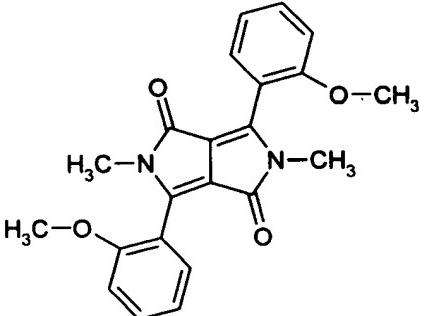
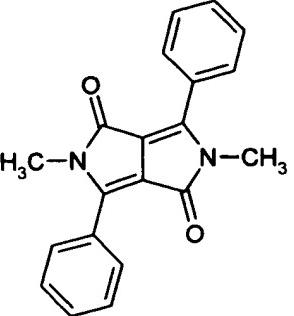
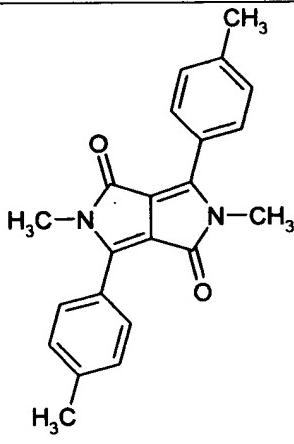
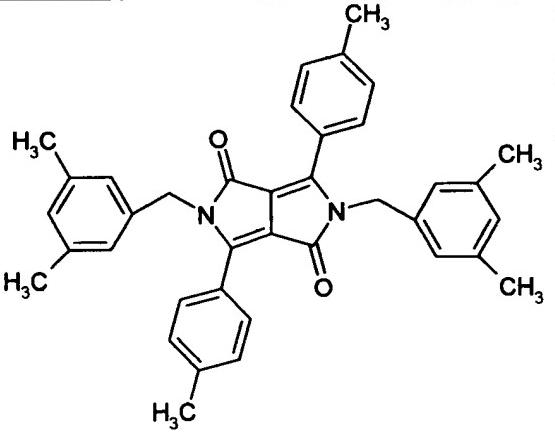
I, Yamamoto Hiroshi, hereby declare as follows:

I am a chemist and hold the degree of Master of technology in organic chemistry from Osaka University in Japan. I have worked in the field of organic and inorganic dyestuffs and pigments for thirteen (13) years directing efforts for the research and development of pigments for the electronic application of photoconductors, solar cells and electroluminescent devices and for the mass coloration of paints, lacquers and thermoplastics. Based on my education and experience, I consider myself to be an expert in the field of organic and inorganic pigments and related technology.

I am a co-inventor of the above-identified patent application.

The following Comparative Examples were conducted by me or under my supervision.

**Comparative Examples 1 to 3**

 <p><b>Comparative Compound C-1</b> (EP-A-499,011: Example 1)</p>	 <p><b>Comparative Compound C-2</b> (EP-A 499,011: Example 7; Compound 2a of DE-A-3713459)</p>
 <p><b>Comparative Compound C-3</b> (EP 0 133 156 A: Example 6)</p>	 <p><b>Inventive Compound I-1</b> (Example 19 of the present application)</p>

**Comparative Example 1:**

Compound C-1, Compound C-2, Compound C-3 and the compound of Example 19 of the present application (I-1) are employed to form evaporated thin films on glass substrates. The film thickness is 60 nm, respectively. The films are then kept in ambient atmosphere. In order to evaluate the morphological stability of the compounds, optical densities of the films are measured using HITACHI U-3300 spectrophotometer. The results are summarized in the table 1 below.

**Table 1**  
**Modulation of optical properties of the samples (OD: Optical Density)**

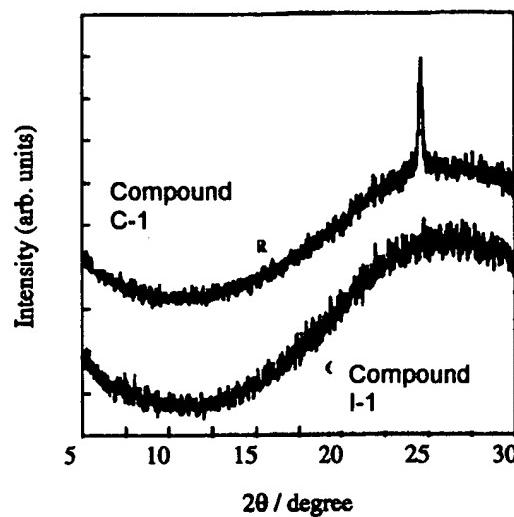
Time	I-1		C-1		C-2		C-3	
	OD	$\lambda_{\max}$ (nm)	OD	$\lambda_{\max}$ (nm)	OD	$\lambda_{\max}$ (nm)	OD	$\lambda_{\max}$ (nm)
0 day	0.55	496	0.44	460	0.32	495	0.36	497
after 6 days	0.55	496	0.41	459	0.25	495	0.30	528
after 20 days	0.55	496	-	-	-	-	-	-

The optical density of C-1, C-2 and C-3 decreases after 6 days from the film formation. This implies that the molecule begins crystal growth on the substrate, thereby optical thickness of the sample increases. In fact the film becomes opaque. Particularly the absorption maximum wavelength of Compound C-3 red-shifts by ca. 30nm. This obviously suggests that the cohesive state of the compound drastically changed.

The optical density and absorption maximum wavelength of I-1 can be regarded as unchanged even after 20 days from the film fabrication. All of the above demonstrate that the morphological stability of I-1 in evaporated films is superior to that of Compounds C-1, C-2 and C-3.

#### **Comparative Example 2:**

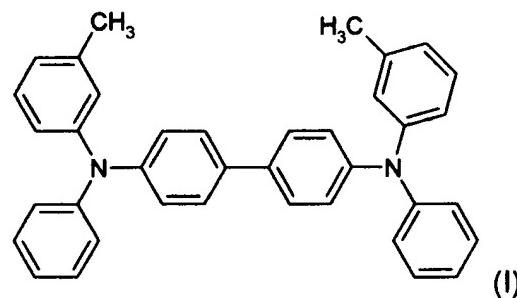
The crystallinity of Compound C-1 and C-2 is already described in D-1. In order to study the degree of crystallinity of the other compounds of Comparative Example 1, the thin films of Compound C-3 (after 6-days) and I-1 (after 20-days) from the film fabrication were employed for XRD measurement. Figure 2 shows the XRD charts obtained. The chart of Compound C-3 displays one reflection peak, indicating that the film is in crystalline state. The chart of I-1 clearly suggests that the film is completely amorphous even after 20 days from the fabrication.



### **Comparative Example 3:**

Morphological smoothness and stability are crucial for thin film type organic EL devices. Namely the compound property to form homogeneous/uniform/smooth and flat evaporated films is indispensable for the light-emitting substances for organic EL devices in order not to produce pinholes or any other defects. If the film obtained is optically heterogeneous/uneven/rough i.e. visually opaque, these morphological properties may cause leakage current in the device operation and consequently the device cannot emit efficiently electroluminescence. This point is experimentally proved as follows.

The above four compounds are applied for the light-emitting substance of the device.



On an ITO glass substrate (made by Geomatech Co. Ud., ITO film thickness 200 nm, sheet resistance  $10 \Omega/\text{cm}^2$ ), Compound (I) represented by the above formula is deposited as a hole transporting substance by vacuum evaporation under a vacuum

of  $5.0 \times 10^{-6}$  Torr and at a depositing rate of 0.05 nm/sec to a membrane thickness of 50 nm to form a hole transporting layer.

Then, on the hole transporting layer thus prepared, Compound C-1, C-2, C-3 or I-1 is deposited as a light-emitting material under a depositing condition of  $5.0 \times 10^{-6}$  Torr and 0.05 nm/sec to a membrane thickness of 50 nm to form a light-emitting layer. Then, on the light-emitting layer, Mg and Ag are co-deposited (atomic ratio Mg:Ag = ca. 10: 1) on it to a film thickness of 200 nm.

By using the ITO side as the anode and the Mg:Ag side as the cathode, a bias of 13-14V is applied to the above element. The devices employing Compound C-1 and I-1 indicate electroluminescence. The device performances are summarized in Table 2 below. As tabulated, the EL emission efficiency (cd/A) of Compound I-1 is 5 times more efficient than that of Compound C-1. Compounds C-2 and C-3 showed no electroluminescence.

The emission from the device using Compound C-1 is heterogeneous/uneven on the emission area of the device as confirmed in Comparative Example 1 (Table 8). This is due to uneven/rough surface morphology of the evaporated film of Compound C-1. On the other hand, the devices employing Compound I-1 exhibit aesthetically beautiful and homogeneous/uniform EL emission on the emission area of the device. This is due to morphological homogeneity of the evaporated film.

**Table 2**

Comparison of organic EL device performances

Sample ID	Bias voltage (V)	Current (mA/cm <sup>2</sup> )	Intensity (cd/m <sup>2</sup> )	Efficiency (cd/A)
I-1	14	410	824	0.20
C-1	13	729	322	0.04

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment,

or both under Section 1001 of Title 18 of the United States Code and that such wilful false statements may jeopardise the validity of this application or any patent issuing thereon.

*April 19th, 2002*

Signed, this day    of April, 2002



Yamamoto Hiroshi